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NONDESTRUCTIVE NEUTRON AND GAMMA-RAY TECHNOLOGIES APPLIED TO GNEP AND SAFEGUARDS

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ABSTRACT

In recent years, LLNL has developed methods for diagnosing significant quantities of special nuclear material (SNM). Homeland security problems have recently focused our attention on detection of shielded highly enriched uranium (HEU), which is a weak signal problem. Current and advanced safeguards applications will require working in the opposite extreme of strong but buried signals. We will review some of the technologies that have been developed at LLNL for homeland security applications and discuss how they might be used in support of international safeguards.

INTRODUCTION

The nuclear fuel cycle of the future presents many technological challenges both in the near term, to determine the expected characteristics of the spent fuel and process monitoring, and in the long term, safeguards. A specific challenge is how much fissionable material is present in a particular sample, production line or storage area. At different stages in the processing, how much SNM (^{235}U and $^{239}\text{Pu}/^{241}\text{Pu}$) is left in the spent fuel from the Advanced Fuel Cycle Facility? Is the expected amount of SNM in the production line, storage area, or waste lines? How can we prove that none of the SNM has been diverted? In this paper we will discuss two new technologies developed at LLNL that could be applied to safeguards and GNEP. These are: 1) fast neutron counting with automated analysis, and 2) compact solid-state thermal neutron detectors. New analysis methods will be briefly discussed.

FAST NEUTRON COUNTING

LLNL has developed a segmented liquid scintillator multiplicity counter with nanosecond timing, which has 10^{-5} discrimination of neutrons and gamma-rays above 500 keV. This detector is modular and scalable in size depending on overall efficiency requirements. It is based on NE-213 xylene-based liquid scintillator modules, now available as BC-501A from Bicron and EJ-301 from Eljen. A photo of a 4" x 3" EJ-301 xylene liquid scintillator detector and PMT housing is shown in Figure 1. This type of liquid scintillator can also be used for neutron spectroscopy by unfolding the energy spectrum using a set of response functions. Historically liquid scintillator detectors have had relatively poor gamma ray – neutron discrimination from analogue pulse-shape discrimination (PSD). With fast digital electronics, far superior neutron – gamma ray identification and much lower energy thresholds yield much better overall neutron

efficiency and identification than is possible with analogue PSD's. The fast digital electronic DAQ system can handle list mode data readout $\sim 10\text{KHz/Ch.}$ (see Figure 2.)



Figure 1. Photograph of a 4" diameter x 3" Eljen EJ-301 xylene based liquid scintillator detector with integration PMT.

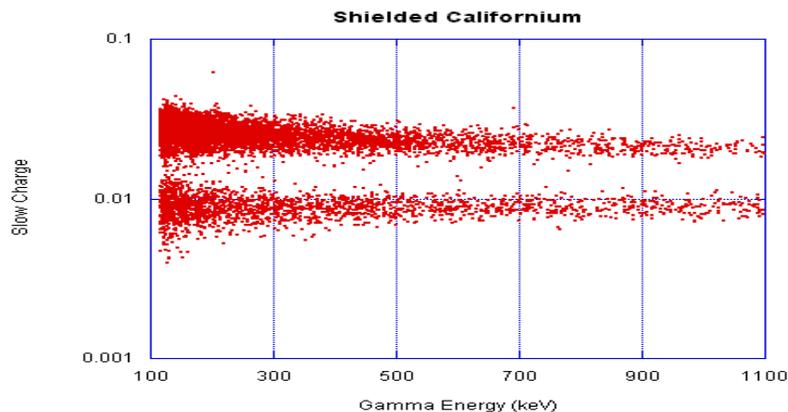


Figure 2. Separation between neutrons and gamma-rays in the DAQ system.

Used passively, this fast neutron counter can detect isolated fission chain bursts in the presence of high backgrounds. High background may occur in storage areas, or when the signal is dominated by (alpha,n) or additional spontaneous fission sources. With ^3He detectors, neutrons created in isolated bursts are spread in time by neutron diffusion times required to thermalize the neutrons. Consequently neutrons from different spontaneous fission events overlap in time, making it difficult to separate the contributions of multiple kinds of neutron sources. This is especially true when the different sources are qualitatively statistically similar, as they are for different spontaneous fission sources. With this liquid scintillator detector array we have recently demonstrated the isolation of individual fission chains in low-multiplying Pu. The liquid scintillator array can be useful in quantifying total fissile mass by isolating spontaneous fission bursts; statistically the different distributions of emitted neutrons (and gamma-rays) from Cm versus Pu

spontaneous fission could be distinguished, and the absolute masses determined (from the rates of the separate kinds of spontaneous fission sources).

Used actively in conjunction with 60 keV neutron interrogation, the liquid scintillators are threshold neutron detectors, blind to the interrogating beam, but responding to the induced high-energy fission neutrons. The 60 keV beam energy selectively fissions ^{235}U and ^{239}Pu and not ^{238}U or ^{232}Th . With fast timing to isolate induced fission events, this technology could be useful for high count rate applications.

This detector array represents two of the suggested highest payoff technologies for NDA safeguards for GNEP: Passive Neutron Albedo Reactivity and Liquid Multiplicity counter with list mode data acquisition¹.

ANALYSIS METHODS

We have developed at LLNL a complete analytic point model theory of neutron counting distributions and time interval distributions. The foundation of the theory is a complete analytic solution² of the Bohnel³ fission chain model. The fission chain theory completes the Dierckx and Hage⁴ and Hage and Cifarelli⁵ theory of random time gate and triggered time gate counting distributions. We have also developed a theory of time interval distributions, including skipped intervals⁶. Using this theory we perform a multi-parameter maximum-likelihood analysis to determine the parameters values that characterize the distributions^{7,8}. The parameters are: the system multiplication, a diffusion time constant (determined from the time-gate dependent Feynman variance to mean – see *Figure 3*), detection efficiency, spontaneous and (alpha, n) source strengths, external direct shine Poisson-distributed background count rate (that can also induce fission, as in active interrogation with an (alpha, n) source). A provisional model of cosmic ray background is also included. The theory has also been extended to include the neutron capture gamma-ray signal.

A random source used in active interrogation of HEU, and that dominates the count rate due to direct shine on the ^3He counters, is robustly separated in this analysis. This is because the Poisson contribution is qualitatively different from the fission signal, and its contribution is characterized by only a single parameter, the external count rate. This separation of sources capability can possibly be extended to the problem of separating the Pu contribution from a dominant Cm source, especially if this analysis is applied to the fast timing signal.

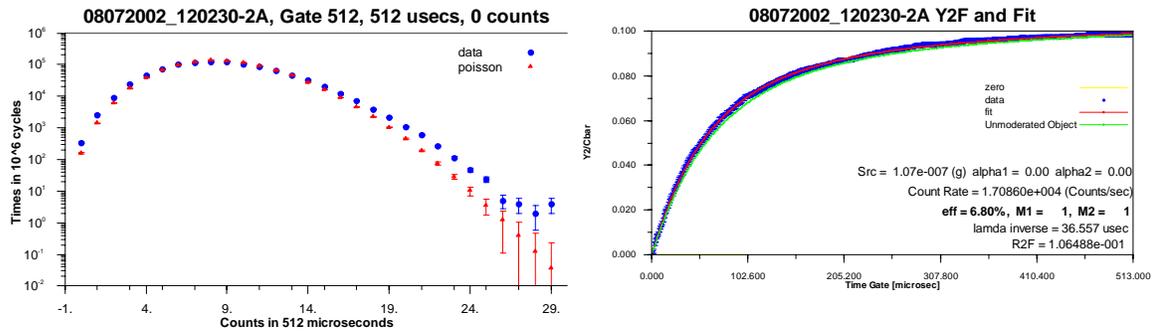


Figure 3. ²⁵²Cf spontaneous fission count distribution (blue) compared to a Poisson distribution of the same count rate (red) for a 512 μsec time gate. The right panel shows the time dependent Feynman variance to mean. The last point is determined by the increased variance of the count distribution in the left panel.

The liquid scintillator detectors also bring energy discrimination of the neutron flux which will inherently contain some information of the source (spontaneous fission vs. alpha induced neutrons) but also leads to our third area of investigation which is low energy neutron interrogation of a sample. SNM has the feature of significant fast neutron induced fission cross-sections in the 10's of keV neutron energy range which is several orders of magnitude greater than that for the Cm isotopes (which are also much scarcer than the SNM isotopes in the expected spent fuel stream). Therefore measuring with the resulting increase in fast neutrons of energy greater than an interrogating beam of 60-100 keV neutrons will determine when compared to the passive flux, the relative amount of SNM in the sample or stream in question.

SOLID STATE NEUTRON DETECTORS

LLNL is currently developing configurable, real-time, low-power, compact, solid state neutron detection system for special nuclear materials neutron signature detection with 10% thermal neutron detection efficiency. This neutron detection system can be used for long term monitoring of nuclear material in storage, or it can be an added feature in a tag or seal. These neutron detection systems can be also used for detecting or monitoring neutron emissions in areas (e.g., underwater, in pipes, etc.) where there are complex scattering environments, no available power, and little or no human access. There are two configurations in the neutron detector, namely ¹⁰B for thermal neutrons and ^{6,7}LiF for higher energy neutrons.

LLNL originally developed novel inexpensive neutron detectors for the long-term monitoring of neutron activity from contraband nuclear material in areas that are inaccessible, such as a cargo-ship container during transport⁹ Our design uses 5mm-diameter, self-biased ¹⁰B-coated neutron detectors (tiny neutron tags) that have 2% efficiency for thermal neutron detection. These detectors have been demonstrated in the laboratory that they can detect neutrons emitted from plutonium in a complex scattering environment in a reasonable amount of time. These coin-sized detectors (when mass-

produced) could be ~\$20 a piece. For Safeguards, a simple detector that can be placed on pipes and casks to determine bulk measurement of neutrons (or lack thereof) for tracking processes and ensuring there is no diversion.

We have developed a prototype of a tiny tag with 15mW electronics (*See Figure 4*). Time information of each event is stored for further data analysis. These semi-conductor detectors are better than currently available ^6Li doped scintillation fibers and photomultiplier based scintillation detectors. They are also immune to gamma-rays and stable in comparison to ^3He tubes.



Figure 4. Solid state based neutron detection system with 15mW electronics.

More recently, we are developing next generation solid-state neutron detectors with thermal neutron detection efficiency beyond 25%. These high aspect ratio pillars¹⁰ will be filled with CVD ^{10}B , $^{6,7}\text{LiF}$ for both thermal and low energy neutron detection. (Figure 5)

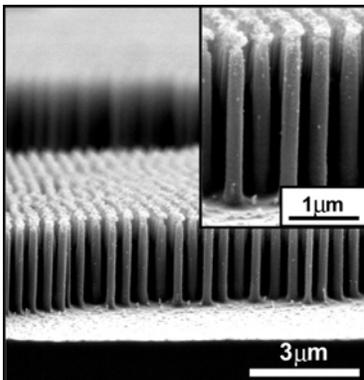


Figure 5. High aspect ratio silicon pillar structure developed at LLNL. When filled with ^{10}B and $^{6,7}\text{LiF}$, the neutron detection efficiency for this system can be as high as 50%.

These inexpensive tiny tags can be used for ubiquitous neutron monitoring in the GNEP processes as “smart tags” for tracking material flow. In a centrifuge facility, they may be used as neutron imagers within the centrifuge hall to monitor for possible diversions (Neutron imagers have been suggested by Mark Pickrell.)¹¹ The information can be propagated with wireless options.

SUMMARY

We have discussed two technologies developed at LLNL for Homeland Security applications. We have given some reasons why these techniques can be brought to bear on the Safeguards problem.

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